

(19)



Europäisches Patentamt
European Patent Office
Office européen des brevets



(11)

EP 0 611 801 B1

(12)

EUROPEAN PATENT SPECIFICATION

(45) Date of publication and mention
of the grant of the patent:
30.07.1997 Bulletin 1997/31

(51) Int Cl. 6: **C08L 23/02, C08F 297/08**

(21) Application number: **94102295.6**

(22) Date of filing: **16.02.1994**

(54) Olefin polymer films

Polyolefin-Filme

Films de polyoléfine

(84) Designated Contracting States:
AT BE CH DE FR GB IT LI NL SE

(74) Representative: **Fuchs, Luderschmidt & Partner**
Patentanwälte
Postfach 46 60
65036 Wiesbaden (DE)

(30) Priority: **17.02.1993 US 18634**

(56) References cited:
EP-A- 0 444 671 **EP-A- 0 490 353**

(43) Date of publication of application:
24.08.1994 Bulletin 1994/34

Remarks:

The file contains technical information submitted
after the application was filed and not included in this
specification

(73) Proprietor: **MONTELL NORTH AMERICA INC.**
Wilmington Delaware 19850-5439 (US)

(72) Inventor: **Giacobbe, James**
Kent County, Worton, Maryland 21678 (US)

EP 0 611 801 B1

Note: Within nine months from the publication of the mention of the grant of the European patent, any person may give notice to the European Patent Office of opposition to the European patent granted. Notice of opposition shall be filed in a written reasoned statement. It shall not be deemed to have been filed until the opposition fee has been paid. (Art. 99(1) European Patent Convention).

Description

This invention relates to thermoplastic films, sheets and laminates and coextruded materials formed therefrom, and films and sheets formed from blends of an olefin polymer composition applied to a base film or sheet of a metallic substrate or of different olefin polymer materials.

In many film applications, such as packaging of foodstuffs, chemical and hazardous materials and in medical applications, the industry requires films having certain properties. In the packaging of foodstuffs, for example, the films must have high puncture resistance, high clarity and gloss, and reduced permeability to gases and/or vapors. The films used to manufacture containers for chemicals and hazardous waste materials must have a high puncture resistance, high elongation strength, high tear resistance and chemical resistance. Films used in medical applications, such as blood bags, must have a high puncture resistance, low modulus, high tear resistance and auto-clavability.

Thus, there is a need for a polymer material which has a lower flexural modulus, good tear strength, higher elastic recovery, reduced draw resonance, as well as all of the other desirable properties.

Films made from ethylene polymers, homopolymers, e.g., HDPE and LLDPE, and copolymers, e.g., LLDPE, and propylene polymers, such as crystalline homopolymers of propylene and random copolymers of propylene and ethylene, do not provide such a combination of desirable properties.

Attempts have been made to overcome the shortcomings of these polymers by preparing heterophasic mixtures of crystalline propylene polymers and 8 to 25% of an elastomeric propylene-ethylene copolymer by sequential polymerization in the presence of a stereospecific Ziegler-Natta type catalyst. However, films of such heterophasic compositions are subject to the formation of fisheyes, inadequate tear strength or the formation of rough surfaces.

EP-A 0 490 353 discloses a blend comprising:

(a) an isotactic polypropylene matrix containing an ethylene/propylene copolymer dispersed phase, said combination having dual melting peaks, a first peak in the range of 150 °C to 165 °C, attributable to the matrix, and a second peak in the range of about 123 °C to 132 °C having at least 2.2 calories per gram melting endotherm, attributable to the copolymer, component (a) being present in the blend in an amount of 60 to 90 percent by weight based on the weight of the blend; and

(b) an ethylene/1-butene copolymer containing 85 to 90 percent by weight ethylene; having a density in the range of 0.890 to 0.910 gram per cubic centimeter; and having a Mw/Mn ratio of up to 8, component (b) being present in the blend in an amount of 10 to 40 percent by weight based on the weight of the blend.

EP-A 0 444 671 discloses films and laminates which are prepared by known filming or calendering processes using heterophasic propylene polymer compositions comprising:

A) 10-60 parts by weight of homopolymer polypropylene having an isotactic index greater than 90, or crystalline copolymer of propylene with ethylene and/or other alpha-olefins containing more than 85% by weight of propylene and having an isotactic index greater than 85;

B) 10-40 parts by weight of a copolymer containing prevailingly ethylene, which is insoluble in xylene at room temperature;

C) 30-60 parts by weight of an amorphous ethylene-propylene copolymer, soluble in xylene at room temperature and containing 40-70% by weight of ethylene;

said propylene polymer compositions having a ratio between the intrinsic viscosities, in tetrahydronaphthalene at 135 °C, of the portion soluble in xylene and of the portion insoluble in xylene at room temperature of from 0.8 to 1.2.

It has now been found that compositions having low modulus, good tear strength, higher elastic recovery, reduced draw resonance, and retention of all of the other desirable properties can be obtained by blending ethylene copolymers with a heterophasic olefin polymer composition.

Accordingly, this invention provides a thermoplastic film having the desired properties or sheet material comprising a blend of 1) a heterophasic olefin polymer composition which is comprised of:

(a) from 10 to 50 parts of a propylene homopolymer having an isotactic index greater than 80, or a copolymer selected from the group consisting of (i) propylene and ethylene, (ii) propylene, ethylene and a $\text{CH}_2=\text{CHR}$ alpha-olefin, where R is a C_{2-8} straight or branched alkyl, and (iii) propylene and an alpha-olefin as defined in (a) (ii), said copolymer containing over 80% propylene and having an isotactic index greater than 80;

(b) from 5 to 20 parts of a semi-crystalline, essentially linear copolymer fraction having a crystallinity of 20 to 60% by differential scanning calorimetry (DSC), wherein the copolymer is selected from the group consisting of (i) ethylene and propylene containing over 55% ethylene, (ii) ethylene, propylene, and an alpha-olefin as defined in (a) (ii) containing from 1 to 10% of the alpha-olefin and over 55% of both ethylene and alpha-olefin, and (iii) ethylene

and an alpha-olefin as defined in (a) (ii) containing over 55% of said alpha-olefin, which copolymer is insoluble in xylene at room or ambient temperature; and

(c) from 40 to 80 parts of a copolymer fraction wherein the copolymer is selected from the group consisting of (i) ethylene and propylene wherein the copolymer contains from 20% to less than 40% ethylene and, (ii) ethylene, propylene, and an alpha-olefin as defined in (a) (ii) wherein the alpha-olefin is present in an amount of 1 to 10% and the amount of ethylene and alpha-olefin present is from 20% to less than 40%, and (iii) ethylene and an alpha-olefin as defined in (a) (ii) containing from 20% to less than 40% of the alpha-olefin, and optionally with 0.5 to 10% of a diene, said copolymer fraction being soluble in xylene at ambient temperature, and having an intrinsic viscosity of from 1.5 to 4.0 dl/g;

10 with the total of the (b) and (c) fractions, based on the total olefin polymer composition, being from 50% to 90%, and the weight ratio of (b)/(c) being less than 0.4; and

2) a copolymer of ethylene with a $\text{CH}_2=\text{CHR}$ alpha-olefin, where R is a C_{1-8} straight or branched alkyl, having a density of 0.875 g/cm^3 or greater.

15 In another embodiment this invention provides film or sheet article comprising a base film or sheet. A layer of the blend of this invention is applied to at least one surface of a thermoplastic film or a nonwoven material or a metallic substrate.

20 The films made from the blends of the present invention can be used in diapers, especially diaper cover stocks, especially bonded to scrim. In general, films from the blend of the invention can be used in personal care products, e. g. diapers and pull-ups; adult incontinence; disposable medical wear, such as gowns; shoe fascias; and feminine hygiene products.

25 All parts and percentages used in this application are by weight unless otherwise specified. Ambient or room temperature is approximately 25°C .

Component 1) (a) is preferably present in an amount from 10 to 40 parts, most preferably from 20 to 35 parts.

30 When (a) is a propylene homopolymer, the isotactic index is preferably from 85 to 98. When (a) is a copolymer, the amount of propylene in the copolymer is preferably from 90 to 99%.

Component 1) (b) is preferably present in an amount from 7 to 15 parts. The crystallinity is 20 to 60% by differential scanning calorimetry (DSC). Generally, the ethylene or said alpha-olefin content or the combination of ethylene and said alpha-olefin when both are used is over 55% up to 98%, preferably from 80 to 95%.

35 Component 1) (c) is preferably present in an amount from 50 to 70 parts. The ethylene or said alpha-olefin content or ethylene and said alpha-olefin content of component (c) is preferably from 20 to 38%, most preferably from 25 to 38%. When component (c) is a terpolymer, the said alpha-olefin is typically present in an amount from 1 to 10%, preferably 1 to 5%. The preferred intrinsic viscosity is from 1.7 to 3 dl/g.

40 The total amount of 1) (b) and (c), based on the total olefin polymer composition is preferably from 65 to 80% and the weight ratio of (b)/(c) is preferably from 0.1 to 0.3.

The total content of copolymerised ethylene or said alpha-olefin units, or of ethylene and said alpha-olefin units when both are present, in component 1) of the blend of this invention is from 15% to 35% by weight.

45 In addition, the compositions of component 1) have a flexural modulus of less than 150 MPa, generally from 20 and 100 MPa; a tensile strength at yield of from 10 to 20 MPa, elongation at break over 400%; a tension set, at 75% strain, from 20% to 50%; a Shore D hardness from 20 and 35; and do not break (no brittle impact failure) when an IZOD impact test is conducted at -50°C .

50 Component 2) is a copolymer of ethylene with a $\text{CH}_2=\text{CHR}$ alpha-olefin, where R is a C_{1-8} , preferably a C_{2-6} , and most preferably a C_{4-6} straight or branched alkyl. The alpha-olefin is present in an amount from 1 to 10%, and preferably from 6-10%. Suitable ethylene copolymers useful as component 2) include ethylene/butene-1, ethylene/4-methyl-1-pentene, ethylene/hexene-1 and ethylene/octene-1. The copolymer can be a LLDPE, or VLDPE, preferably a LLDPE, where the comonomer is 1-octene. Preferably the density of the ethylene copolymer is 0.890 to 0.940 g/cm^3 , and most preferably from 0.890 to 0.927 g/cm^3 .

55 The blends of the present invention contain from 60 to 95%, by weight, of component 1) and from 5 to 40%, by weight, of component 2). Preferably, component 1) is present in an amount of from 90 to 75% and component 2) is present in an amount of from 10 to 25%.

The blends of this invention have at least one melt peak, determined by DSC, present at temperatures higher than 120°C , and at least one peak, relative to the vitreous transition, present at temperatures from -10°C and -35°C .

60 Typically, the blends of the present invention have a flexural modulus of less than 150 MPa, generally from 20 to 100 MPa.

65 Copolymers and terpolymers of propylene and ethylene or an alpha-olefin or of propylene, ethylene and an alpha-olefin are preferred as component 1) (a), and copolymers of propylene with ethylene or an alpha-olefin are most preferred as component (a).

70 Suitable alpha-olefins of the formula $\text{CH}_2=\text{CHR}$ include butene-1, pentene-1, 4-methylpentene-1, hexene-1, and

octene-1. When used to prepare component 1) (a) they are present in such quantities that the isotactic index of the resulting polymer is not less than 80%.

When a diene is present during the preparation of components 1) (b) and (c), it is typically a butadiene, 1,4-hexadiene, 1,5-hexadiene, ethylidene norbornene diene monomer and is typically present in amount from 0.5 to 10%, preferably 1 to 5%.

The component 1) can be prepared with a polymerization process comprising at least two stages, where in the first stage the propylene or propylene and ethylene or said alpha-olefin or propylene, ethylene or said alpha-olefin are polymerized to form component 1) (a), and in the following stages the mixtures ethylene and propylene or said alpha-olefin or ethylene, propylene and said alpha-olefin, and optionally a diene, are polymerized to form components 1) (b) and (c).

The polymerization can be conducted in liquid phase, gas phase, or liquid-gas phase using separate reactors, all of which can be done either by batch or continuously. For example, it is possible to carry out the polymerization of component 1) (a) using liquid propylene as diluent, and the polymerization of components 1) (b) and (c) in gas phase, without intermediate stages except for the partial degassing of the propylene. This is the preferred method.

The polymerization reactions are carried out in an inert atmosphere in the presence of an inert hydrocarbon solvent or of a liquid or gaseous monomer.

Suitable inert hydrocarbon solvents include saturated hydrocarbons, such as propane, butane, hexane and heptane.

Hydrogen can be added as needed as a chain transfer agent for control of the molecular weight.

The reaction temperature in the polymerization of component 1) (a) and for the polymerization of components 1) (b) and (c), can be the same or different, and is generally from 40°C to 90°C, preferably 50 to 80°C for the polymerization of component 1) (a), and 40 to 65°C for the polymerization of components 1) (b) and (c).

The pressure of the polymerization of component 1) (a), if carried out in liquid monomer, is the one which competes with the vapor pressure of the liquid propylene at the operating temperature used, eventually modified by the vapor pressure of the small quantity of inert diluent used to feed the catalyst mixture, and the overpressure of optional monomers and the hydrogen used as molecular weight regulator.

The pressure of the polymerization of components 1) (b) and (c), if done in gas phase, can be from 506625 to 3039750 Newtons/m² (5 to 30 atm). The residence times relative to the two stages depend on the desired ratio between fraction (a) and (b) + (c), and are usually from 15 min. to 8 hours.

The catalyst used in the polymerization comprises the reaction product of 1) a solid component containing a halogen-containing titanium compound and an electron-donor compound (internal donor) supported on an activated magnesium chloride, 2) a non-halogen containing Al-trialkyl compound and 3) an electron-donor compound (external donor).

Suitable titanium compounds include those with at least one Ti-halogen bond, such as halides and alkoxy halides of titanium.

In order to obtain these olefin polymer compositions in the form of flowable spherical particles having a high bulk density, the solid catalyst component must have a) a surface area smaller than 100 m²/g, preferably between 50 and 80 m²/g, b) a porosity from 0.25 to 0.4 cc/g. and c) an X-ray spectrum, where the magnesium chloride reflections appear, showing the presence of a halo between the angles 2 θ of 33.5° and 35° and by the absence of the reflection at 2 θ of 14.95°. The symbol θ = Bragg angle.

The solid catalyst component is prepared by forming an adduct of magnesium dichloride and an alcohol, such as ethanol, propanol, butanol and 2-ethylhexanol, containing generally 3 moles of alcohol per mole of MgCl₂, emulsifying the adduct, cooling the emulsion quickly to cause the adduct to solidify into spherical particles, and partially dealcoholating the particulate adduct by gradually increasing the temperature from 50°C to 130°C for a period of time sufficient to reduce the alcohol content from 3 moles to 1-1.5 moles per mole of MgCl₂. The partially dealcoholated adduct is then suspended in TiCl₄ at 0°C, such that the concentration of adduct to TiCl₄ is 40-50 g/l TiCl₄. The mixture is then heated to a temperature of 80°C to 135°C for a period of 1-2 hr. When the temperature reaches 40°C, sufficient electron donor is added so that the desired molar ratio of Mg to electron donor is obtained.

An electron-donor compound selected preferably among the alkyl, cycloalkyl, and aryl phthalates, such as for example diisobutyl, di-n-butyl, and di-n-octyl phthalate, is added to the TiCl₄.

When the heat treatment period has ended, the excess hot TiCl₄ is separated by filtration or sedimentation, and the treatment with TiCl₄ is repeated one or more times. The solid is then washed with a suitable inert hydrocarbon compound, such as hexane or heptane, and dried.

The solid catalyst component typically has the following characteristics:

Surface area	less than 100 m ² /g, preferably between 50 and 80 m ² /g
Porosity	0.25 - 0.4 cc/g

(continued)

5	Pore volume distribution X-ray spectrum	50% of the pores have a radius greater than 10 nm (100 angströms). where the Mg chloride reflections appear, showing a halo with maximum intensity between angles of 2 θ of 33.5° and 35°, and where the reflection of 2 θ of 14.95° is absent.
---	--	--

The catalyst is obtained by mixing the solid catalyst component with a trialkyl aluminum compound, preferably triethyl aluminum and triisobutyl aluminum, and an electron-donor compound.

10 Various electron donor compounds are known in the art. The preferred electron donor compounds are those silane compounds having the formula R'R"Si(OR)₂ where R' and R" may be the same or different and are C₁₋₁₈ normal or branched alkyl, C₅₋₁₈ cycloalkyl, or C₆₋₁₈ aryl radicals, and R is a C₁₋₄ alkyl radical.

15 Typical silane compounds which may be used include diphenyldimethoxysilane, dicyclohexyldimethoxysilane, methyl-t-butylidimethoxysilane, diisopropylidimethoxysilane, dicyclopentyldimethoxysilane, cyclohexylmethyldimethoxysilane and phenyltrimethoxysilane.

The Al/Ti ratio is typically between 10 and 200 and the Al/silane molar ratio between 1/1 and 1/100.

The catalysts may be precontacted with small quantities of olefin monomer (prepolymerization), maintaining the catalyst in suspension in a hydrocarbon solvent and polymerizing at a temperature from room temperature to 60°C for a time sufficient to produce a quantity of polymer from 0.5 to 3 times the weight of the catalyst.

20 This prepolymerization also can be done in liquid or gaseous monomer to produce, in this case, a quantity of polymer up to 1000 times the catalyst weight.

The content and amount of catalyst residue in the thermoplastic olefin polymers of this invention is sufficiently small so as to make the removal of catalyst residue, typically referred to as deashing, unnecessary.

25 The heterophasic olefin polymer, component 1), prepared with the aforementioned catalyst are in spheroidal particle form, and the particles have a diameter from 0.5 to 7 mm.

The heterophasic olefin polymer used in the blends of this invention can be a "visbroken" polymer prepared from spherical particles as described above, having a melt flow rate (MFR, according to ASTM D-1238, measured at 230°C, 2.16 kg) of from 5 to 400, preferably from 10 to 200, and most preferably from 20 to 100, from an initial MFR of from 0.2 to 20, and preferably 0.5 to 3.

30 Alternatively, component 1) can be produced directly in the polymerization reactor to the preferred MFR.

The process of visbreaking component 1) is well known in the art. Generally, it is carried out as follows: heterophasic olefin polymer material in "as polymerized" form, e.g., flaked, powders or spheres out of the polymerization reactor or pelletized, has sprayed thereon or blended therewith, a prodegradant or free radical generating source, e.g., a peroxide in liquid or powder form or a peroxide/polypropylene concentrate, such as @Xantrix 3024 peroxide concentrate available from HIMONT U.S.A., Inc. The heterophasic olefin polymer material and peroxide is then introduced into means for thermally plasticizing and conveying the mixture, e.g., an extruder at elevated temperature. Residence time and temperature are controlled in relation to the particular peroxide selected (i.e., based on the half-life of the peroxide at the process temperature of the extruder) so as to effect the desired degree of polymer chain degradation. The net result is to narrow the molecular weight distribution of the polymer as well as to reduce the overall molecular weight and thereby increase the MFR relative to the as-polymerized polymer. For example, a polymer with a fractional MFR (i.e., less than 1), or a polymer with a MFR of 0.5 to 10, can be selectively visbroken to a MFR of 15 to 50, preferably 28 to 42, by selection of peroxide type, extruder temperature and extruder residence time without undue experimentation. Sufficient care should be exercised in the practice of the procedure to avoid crosslinking in the presence of an ethylene-containing copolymer; typically, crosslinking can be easily avoided where the ethylene content of the copolymer is sufficiently low.

45 The rate of peroxide decomposition is defined in terms of half-lives, i.e., the time required at a given temperature for one-half of the peroxide molecules to decompose. It has been reported (U.S. 4,451,589) for example, that using @Lupersol 101 peroxide under typical extruder pelletizing conditions 232 °C (450°F), 2 1/2 minutes residence time), only 2 x 10⁻¹³ of the peroxide would survive pelletizing.

50 In general, the prodegradant should not interfere with or be adversely affected by commonly used polypropylene stabilizers and should effectively produce free radicals that upon decomposition initiate degradation of the polypropylene moiety. The prodegradant should have a short enough half-life at a polymer manufacturing extrusion temperatures, however, so as to be essentially entirely reacted before exiting the extruder. Preferably, they have a half-life in the polypropylene of less than 9 seconds at 288°C (550°F) so that at least 99% of the prodegradant reacts in the molten polymer before 1 minute of extruder residence time. Such prodegradants include, by way of example the following: 2,5-dimethyl-2,5-bis(t-butylperoxy) hexane-3 and 4-methyl-4-t-butylperoxy-2-pentanone (e.g. @Lupersol 130 and @Lupersol 120 peroxides available from Lucidol Division Penwalt Corporation); 3,6,6,9,9-pentamethyl-3-(ethylacetate)-

1,2,4,5-tetraoxycyclononane (e.g., USP-138 peroxide from Witco Chemical Corporation); and 1,1'-bis(tert-butylperoxy) diisopropylbenzene (e.g., ®Vulcup R peroxide from Hercules Incorporated). Preferred concentrations of the free radical source prodegradants are in the range of from 0.01 to 0.4 percent based on the weight of the polymer. Particularly preferred is ®Lupersol 101 peroxide

5 The blends of the invention can be prepared by mechanically blending component 1) and component 2) by conventional mixing processes, in conventional compounding equipment.

Unless otherwise specified, the following analytical methods are used to characterize the supported catalyst component, the heterophasic olefin polymer compositions, films prepared therefrom and comparative film materials.

10	Properties	Method
	Melt Flow Rate, g/10 min.	ASTM-D 1238, condition L
	Ethylene, wt %	I. R. Spectroscopy
15	Intrinsic viscosity	Determined in tetrahydronaphthalene at 135°C
	Xylene solubles, wt %	See description below.
	Flexural modulus at 23°C and Vitreous transition temperature	Using a device for dynamic-mechanical measurements of DMTA of Polymer Laboratories at a frequency measure of 1 Hz and a scanning temperature of 2°C/min. A sample plaque (40x10x2 mm) of the polymer to be analyzed is cut from a pressure molded sheet prepared with a Carver press at 200°C with 9071.847 kg (10 tons) of pressure for 10 minutes and then cooling the sheet at 15°C/min.
20		
	Tension set at 75%	ASTM-D 412
25	Tensile Strength at yield and at break	ASTM-D 638
	Elongation at yield and at break	ASTM-D 638
	Surface area	B.E.T.
	Porosity	B.E.T.
30	Bulk density	DIN-53194
	Elemendorf tear	ASTM-D 1922-78
	Dart impact strength	ASTM D 4272-83

35 Unless otherwise specified, the compositions of the present invention are produced by a general procedure comprising tumble blending component 1), which has been visbroken with ®Lupersol 101, 2,5-dimethyl-2,5-bis(t-butylperoxy) hexane, and component 2) set forth below in the Examples. Samples of the blend to be subjected to the various physical-mechanical analyses are molded by use of a Negri & Bossi 90 injection press, after stabilizing the material with 0.05 pph ®Cyanox 1790 and 0.05 pph calcium stearate, and pelletizing it with a single screw Bandera extruder (cylinder diameter 30 mm) at 210°C. The analytical conditions are as follows:

40 temperature of the melt 250°C
 temperature of the mold 60°C
 injection time 20 sec.
 cooling time 25 sec.

45 The samples of the film materials were 20.32 to 55.88 µm (0.8 to 2.2 mil) in thickness and were cut from the film sheet in the size provided in the particular ASTM test method being used.

50 The weight percentage of the sum of the components 1) (b) and (c) fractions, indicated by % (b) + (c), is calculated by determining the weight of the mixture fed during the second stage, and comparing it with the weight of the final product.

The weight percentage (%) of the components 1) (a), (b), and (c) fractions described herein are determined as follows:

$$55 \% (a) = 100\% - [(b) + (c)]$$

$$\% (c) = S_f - P_a S_a$$

where S_f and S_a are the percentage by weight of the portion soluble in xylene of the final product and the polypropylene fraction (a), respectively; P_a is the weight ratio between said fraction and the final product.

$$\% \text{ (b)} = 100 - \% \text{ (a)} - \% \text{ (c)}$$

The percentage by weight of ethylene or said alpha-olefin or ethylene and said alpha-olefin contained in copolymer fraction component 1) (c) soluble in xylene is calculated using the following formula:

wt.% ethylene and/or said alpha-olefin in fraction (c) = $\frac{(C_f - C_a)}{1 - X} \cdot X$

where:

C_f = % ethylene and/or said alpha-olefin in the xylene solubles of the final product;
 C_a = % ethylene and/or said alpha-olefin in the xylene solubles of fraction (a);
 $X = S_a \cdot P_a / S_f$

20 The intrinsic viscosity of fraction 1) (c), $(I.V.)_{(c)}$, is calculated using the following formula:

$$(I.V._{(c)}) = (I.V._{Sf} - I.V._{(a)} \cdot X) / (1 - X)$$

25 where $I.V_{sf}$ is the intrinsic viscosity of the xylene soluble fraction of the final composition and $I.V_{(a)}$ is the intrinsic viscosity of the xylene soluble portion of component 1) (a) fraction.

The weight percent of component 1) soluble in xylene at room temperature is determined by dissolving 2.5 g of the polymer in 250 ml of xylene in a vessel equipped with a stirrer which is heated at 135°C with agitation for 20 minutes. The solution is cooled to 25°C while continuing the agitation, and then left to stand without agitation for 30 minutes so that the solids can settle. The solids are filtered with filter paper, the remaining solution is evaporated by treating it with a nitrogen stream, and the solid residue is vacuum dried at 80°C until a constant weight is reached. The percent by weight of polymer insoluble in xylene at room temperature is the isotactic index of the polymer. The value obtained in this manner corresponds substantially to the isotactic index determined via extraction with boiling n-heptane, which by definition constitutes the isotactic index of the polymer.

35 Examples illustrative of the component 1), physical properties thereof, a process for preparing same, a film based on blends of said component 1) and component 2) and a method of preparing said film are set forth below.

A) Preparation of $MgCl_2$ /Alcohol Adduct

Under an inert atmosphere, 28.4 g anhydrous $MgCl_2$, 49.5 g of an anhydrous ethanol, 100 ml of ROL OB/30 vaseline oil and 100 ml of silicone oil having a viscosity of $350 \cdot 10^{-6} m^2/s$ (350 cs) are introduced into a reaction vessel equipped with a stirrer and heated at $120^\circ C$ with an oil bath and stirred until the $MgCl_2$ is dissolved. The hot reaction mixture is then transferred under an inert atmosphere to a 1500 ml vessel equipped with an Ultra Turrax T-45 N stirrer and a heating jacket and containing 150 ml of vaseline oil and 150 ml of silicone oil. The temperature is maintained at $120^\circ C$ with stirring for 3 minutes at 3,000 rpm. The mixture is then discharged into a 2 liter vessel equipped with a stirrer containing 1,000 ml of anhydrous n-heptane cooled at $0^\circ C$ with a dry ice/isopar bath and stirred at a tip speed of 6 m/sec for about 20 minutes while maintaining the temperature at $0^\circ C$. The adduct particles thus formed are recovered by filtering, are washed 3 times at room temperature with 500 ml aliquots of anhydrous hexane and gradually heated by increasing the temperature from $50^\circ C$ to $100^\circ C$ under nitrogen for a period of time sufficient to reduce the alcohol content from 3 moles to 1.5 moles per mole of $MgCl_2$. The adduct has a surface area of $9.1 m^2/g$ and a bulk density of 0.564 g/cc.

B) Solid Catalyst Component Preparation

55 The adduct (25 g) is transferred under nitrogen into a reaction vessel equipped with a stirrer and containing 625 ml of $TiCl_4$ at 0°C under agitation. It is then heated to 100°C in 1 hr. When the temperature reaches 40°C, diisobutylphthalate is added in an amount such that the molar ratio of Mg to diisobutylphthalate is 8. The contents of the vessel are heated at 100°C for 2 hours with agitation, the agitation is stopped and the solids are allowed to settle. The hot

liquid is removed by siphon.

550 ml of $TiCl_4$ is added to the solids in the vessel and the mixture heated at 120°C for 1 hr. with agitation. The agitation is stopped and the solids are allowed to settle. The hot liquid is then removed by siphon. The solids are washed 6 times at 60°C with 200 ml aliquots of anhydrous hexane, and then 3 times at room temperature. The solids, after being vacuum dried, have a porosity of 0.261 cc/g, a surface area of 66.5 m²/g and a bulk density of 0.44 g/cc.

Examples 1-3

10 These examples illustrate the heterophasic olefin polymer composition and a method for preparing the polymers.

The polymerization runs are conducted under nitrogen in a 22 liter stainless steel autoclave equipped with a helicoid magnetic stirrer and operated at 90 rpm.

15 All temperatures, pressures and concentrations of olefin monomers and hydrogen, when present, are constant unless otherwise indicated. The concentration of hydrogen and of the relative monomers is analyzed continuously in gas phase with a process gas chromatograph and fed in order to maintain constant the desired concentration of same.

15 The polymerization is a batch process conducted in two stages. The first stage comprising the polymerization of the relevant monomer or monomers in liquid propylene and the second stage the copolymerization of ethylene and propylene in gas phase.

20 In the first stage, the following ingredients in the order in which they are listed are fed into the autoclave at 20°C over a period of 10 minutes: 16 l of liquid propylene, appropriate quantities of ethylene and hydrogen, and the catalyst system consisting of 1) the solid catalyst component (0.15g) prepared as described above, and 2) a mixture of 75 ml of triethyl aluminum (TEAL) at a 10% concentration in hexane and an appropriate quantity of cyclohexylmethyldimethoxysilane (CMMS) electron donor such that the Al/CMMS molar ratio is 7.5. The catalyst system is pressure fed into the autoclave with propylene.

25 The temperature is brought to the desired level in 10 minutes and maintained constant throughout the entire polymerization reaction period. After the established reaction time has passed, essentially all of the unreacted monomer (s) is/are eliminated by degassing at 60°C at essentially atmospheric pressure.

30 In the second stage, the polymer product (a) of the first stage, after taking a sample for the various analyses, is brought to the established temperature for the second stage. Propylene and ethylene are then fed into the autoclave at the ratio and in the quantities established in order to achieve the pressure and the gas phase composition desired.

35 During the polymerization the pressure and gas phase composition are maintained by feeding the propylene and ethylene mixture established by way of instruments which regulate or measure or both regulate and measure the flow rate. The length of the feed varied according to the catalyst system employed and the amount of components 1) b) and c) desired in the particular heterophasic olefin polymer product.

At the end of the second stage polymerization reaction the powder is discharged, stabilized and then oven dried under a nitrogen stream at 60°C.

35 The ingredients and relative operating conditions are set forth in Table IA and the tests results are set forth in Table IB.

Table 1A

40	Examples	1	2	3
		1st Phase		
	Temperature, °C	70	70	70
45	Pressure, kg/cm ² (atm.)	32 (31)	32 (31)	32 (31)
	Time, min.	30	20	30
	H ₂ in gas phase, mol %	0.58	0.10	0.30
	Ethylene in gas phase, mol %	1.45	2.60	2.50
	Ethylene in pol. wt. %	3.0	4.3	4.1
50	Intrinsic Visc., dl/g	2.18	3.09	2.31
	Xylene Sol. (S _a), wt. %	9.4	9.0	0.7
	Ethylene in Xylene Sol. (C _a), wt. %	11	16	17
	Intrinsic Visc. Xylene Sol.(I.V. _a), dl/g	1.15	1.39	1.19
55	2nd Phase			
	Temperature, °C	50	50	50
	Pressure, kg/cm ² (atm.)	11.7 (11.3)	11.9 (11.5)	11.7 (11.3)

Table 1A (continued)

Examples	1	2	3
2nd Phase			
Time, min.	335	500	250
H ₂ in gas phase, mol %	2.23	3.0	2.05
Ethylene in gas phase, mol %	15.9	16.9	22.54

Table 1B

Examples	1	2	3
Final Product			
Yield, Kg Pol/g Cat	11	16.3	9.9
Comonomer, wt. %	24.6	22.7	29.0
Bipolymer (b)+(c), wt. %	70	67	71.8
Intrinsic Visc., dl/g	2.05	2.3	2.34
Xyl. Sol. (S _f), wt. %	63.4	60.5	63.5
Ethylene Xyl. Sol., wt. % (C _f)	30.2	27.0	34.8
Intrinsic Vis. Xyl. Sol. I.V. _{S_f} , dl/g	1.83	2.02	2.12
Fraction (b), wt. %	9.45	9.37	11.34
Fraction (c), wt. %	60.55	57.63	60.46
Ethylene frac. (b), wt. %	51.9	57.1	53.7
Ethylene frac. (c), wt. %	31.1	27.6	35.7
Intrinsic Vis. frac. (c) (I.V. ^c), dl/g	1.86	2.05	2.18
Melt Index, °C	150	147	145
Flexural modulus, MPa	30	77	82
R.C.I. IZOD at -50°C, J/m	NB ¹	NB	NB
Shore D hardness	24	25	20
Tension Set at 75%, %	41	28	36
Tensile strength, MPa	13.8	15.8	15.4
Tensile strength at yield, MPa	5.0	5.8	4.6
Elongation at break, %	517	925	940
Haze, %	31	34	35
Vitreous transition ² , °C	-25(P)	-23(P)	-28(P)
	-75	-119	-81
	-128	-121	-125

¹ NB = no break² (P) = main peak45 Example 4

This example illustrates a cast film material of a blend of the present invention and a method for preparing the same.

A cast film of the blend of the present invention containing 1) 75% of a heterophasic olefin polymer material composition, produced according to the method of Example 2, except that, component 1 (a) is present in an amount of about 37% and 63% is component 1 (b) + (c), visbroken to 30 MFR from an initial, as polymerized MFR of 0.8; and 2) 25% of DOWLEX 2045, a linear low density polyethylene containing octene-1, having a melt index of 1.0 g/10 min and a density of 0.92, g/cm³ is prepared by charging the blend into an extruder, extruding it through a flat film die and quenching onto a chill roll to produce a film of 20.32 µm (0.8 mil) thickness using the following equipment and processing conditions:

55

Screw design:

Compression ratio 4:1 to 2:1

Feed zone depth: 1.10 to 1.24 cm (0.435 to 0.490") (8.89 cm (3.5")

Metering zone depth: 0.32 to 0.36 cm (0.125 to 0.140") for 8.89 cm (3.5") extruder with 3.5:1 compression ratio)
 extruder.

5 Die: Conventional center-fed coathanger manifold.
 Extruder operating conditions: Melt temperature: 204-238°C (400-460°F)
 Extruder Barrel: 177-216°C (350-420° F) from zone 1 to zone 6
 Adapter and die temperatures: 216°C (420°F)

10 Control 1

15 A cast film material of a heterophasic olefin polymer material composition, obtained by sequential polymerization in at least two stages, containing 37% of a propylene-ethylene copolymer, (96.7:3.3 wt. ratio of polymerized units), and 63% of an ethylene-propylene copolymer, (29.7:1 wt. ratio of polymerized units), visbroken to 30 MFR from an initial, as polymerized MFR of 0.8, prepared according to the procedure described above.

Table 2

20	ELONGATION	CONTROL		EXAMPLE 4	
		% DEFORM.	% RECOVERY	% DEFORM.	% RECOVERY
25	10%	4.1	95.9	5.3	94.7
	25%	11.8	88.2	11.8	99.2
	50%	17.0	83.0	15.7	84.4
	75%	20.2	79.8	19.1	81.0

30 As demonstrated in Table 2 above, at 10% and 25% elongation very little difference is seen in the % deformation and recovery between Example 4 and control. However, at elongations of 50% and 75%, in the blend of the present invention, Example 4, the percent deformation decreases and the percent recovery increases, whereas the percent deformation increases and the percent recovery decreases in the composition of the control, which does not contain LLDPE.

35 Illustrated in Table 3 is the tear strength and impact strength of cast film material of the blend of the present invention, Example 4, and the control composition, which does not contain LLDPE.

Table 3

35	PROPERTIES	CONTROL	EXAMPLE 4
	Film Thickness, μm (mil)	55.88 (2.2)	55.8 (2.2)
40	Elmendorf Tear Strength, g (MD/CD)	430/610	440/790
	Dart Impact Strength, g	520	740

50 As demonstrated in Table 3, a significant increase in the tear and impact strength of the blend of the invention is obtained, as compared to the control, which does not contain LLDPE.

55 Various types of film materials of conventional thickness and thin films less than 508 μm (20 mils) thick to as thin as 12.7 μm (0.5 mils) can be prepared using the heterophasic olefin polymer composition described herein as well as heavy film materials, typically referred to as sheets, from 508 to 2540 μm (20 to 100 mils) thick. For example, it can be used to prepare cast films, uniaxially and biaxially oriented films and extruded or calendered sheets. In addition, a layer comprising the heterophasic olefin polymer composition can be applied to, e.g. by lamination, extrusion coating or coextrusion techniques, at least one surface of a thermoplastic film material or a metallic sheet or foil substrate or woven or non-woven material.

55 Typical thermoplastic materials include crystalline homopolymers of a C₂₋₁₀ alpha-olefin monomer, such as propylene or ethylene, or copolymers of propylene with ethylene or with a C₄₋₁₀ alpha-olefin monomer or of propylene with both ethylene and a C₄₋₁₀ alpha-olefin monomer provided that, when the comonomer is ethylene, the maximum polymerized ethylene content is 10%, preferably 4%, and when the comonomer is a C₄₋₁₀ olefin, the maximum polymerized content thereof is 20%, preferably 16%, and when both ethylene and an alpha-olefin are used the maximum polymerized content of both is 30%, preferably 20%, as well as polyesters, polyamides, ethylene-vinyl alcohol copolymers and ethylene-vinyl acetate copolymers. Aluminum is a suitable metallic substrate.

In addition, film materials can be prepared from blends of from 5 to 45% of the present invention described herein

with from 95 to 55% of a crystalline homopolymer of a C₂₋₁₀ alpha-olefin monomer or copolymer of propylene with ethylene or with a C₄₋₁₀ alpha-olefin monomer or of propylene, ethylene and a C₄₋₁₀ alpha-olefin monomer, said copolymer having the maximum polymerized content of ethylene or alpha-olefin or both as described in the preceding paragraph. Preferably, the amount of the blend of the present invention in such blends is from 10 to 30%.

5

Claims

1. A film or sheet material comprising a blend of 1) from 95 to 60%, by weight of a heterophasic olefin polymer 10 composition which is comprised of

(a) from 10 to 50 parts of a propylene homopolymer having an isotactic index greater than 80, or a copolymer selected from the group consisting of (i) propylene and ethylene, (ii) propylene, ethylene and a CH₂=CHR alpha-olefin, 15 where R is a C₂₋₈ straight or branched alkyl, and (iii) propylene and an alpha-olefin as defined in (ii), said copolymer containing over 80% propylene and having an isotactic index greater than 80;
 (b) from 5 to 20 parts of a semi-crystalline, essentially linear copolymer fraction having a crystallinity of 20 to 60% measured by differential scanning calorimetry (DSC), wherein the copolymer is selected from the group 20 consisting of (i) ethylene and propylene containing over 55% ethylene, (ii) ethylene, propylene, and an alpha-olefin as defined in (a) (ii) containing from 1 to 10% of the alpha-olefin and over 55% of both ethylene and alpha-olefin, and (iii) ethylene and an alpha-olefin as defined in (a) (ii) containing over 55% of said alpha-olefin, which copolymer is insoluble in xylene at room or ambient temperature; and
 (c) from 40 to 80 parts of a copolymer fraction wherein the copolymer is selected from the group consisting of 25 (i) ethylene and propylene containing from 20% to less than 40% ethylene, (ii) ethylene, propylene, and an alpha-olefin as defined in (a) (ii) wherein the alpha-olefin is present in an amount of 1 to 10% and the amount of ethylene and alpha-olefin present is from 20% to less than 40%, and (iii) ethylene and an alpha-olefin as defined in (a) (ii) containing from 20% to less than 40% of said alpha-olefin, and optionally with 0.5 to 10% of a diene, said copolymer fraction being soluble in xylene at ambient temperature, and having an intrinsic viscosity of from 1.5 to 4.0 dL/g;

30

with the total of the (b) and (c) fractions, based on the total olefin polymer composition, being from 50% to 90%, and the weight ratio of (b)/(c) being less than 0.4; and

2) from 5 to 40%, by weight of a copolymer of ethylene with a CH₂=CHR alpha-olefin, where R is a C₁₋₈ straight or branched alkyl, having a density of 0.875 g/cm³ or greater.

35

2. The material of claim 1 wherein (a) is a copolymer of propylene and ethylene containing from 90 to 99% propylene.
3. The material of claim 1 wherein (c) is a copolymer of propylene and ethylene containing from 20 to 38% ethylene.
40. 4. The material of claim 1 wherein component (2) is an ethylene copolymer containing an 1-octene comonomer.
5. The material of claim 4 wherein said ethylene copolymer has a density of from 0.890 to 0.927 g/cm³.
6. The material of claim 4 wherein said ethylene copolymer is a linear low density polyethylene.
45. 7. The material of claim 1 wherein component 1) is present in an amount of from 90 to 75% and component 2) is present in an amount of from 10 to 25%.
8. The material of claim 2 or 3 wherein the total content of copolymerized ethylene is from 15% to 35% by weight.
50. 9. A film or sheet article comprising a base film or sheet of a crystalline homopolymer of a C₂₋₁₀ alpha-olefin monomer or of a copolymer selected from the group consisting of (i) propylene with ethylene, (ii) propylene with ethylene and a C₄₋₁₀ alpha-olefin monomer, and (iii) propylene with a C₄₋₁₀ alpha-olefin monomer, provided that, when the comonomer is ethylene, the maximum polymerized ethylene content is 10%, when the comonomer is a C₄₋₁₀ olefin, the maximum polymerized content thereof is 20%, and when both ethylene and a C₄₋₁₀ olefin are used the maximum polymerized content is 30%; and applied to at least one surface of the base film or sheet, a layer of the material of claim 1.

10. A film or sheet article comprising a base film or sheet of a metallic substrate having applied to at least one surface thereof a layer of the material of claim 1 or 7.

11. A nonwoven article having applied to at least one surface thereof a layer of the material of claim 1 or 7.

5

Patentansprüche

1. Ein Material in Form eines Films oder einer Folie enthaltend eine Mischung aus 1) von 95 bis 60 Gew.% einer heterophasischen Olefinpolymer-Zusammensetzung welche enthält:

10 (a) von 10 bis 50 Teilen eines Propylen-Homopolymeren mit einem Isotaktizitäts-Index von mehr als 80, oder ein Copolymer ausgewählt aus der Gruppe bestehend aus (i) Propylen und Ethylen, (ii) Propylen, Ethylen und einem $\text{CH}_2=\text{CHR}$ alpha-Olefin, worin R ein geradkettiges oder verzweigtes C_{2-8} Alkyl bedeutet, und (iii) Propylen und einem alpha-Olefin, wie in (ii) definiert, wobei besagtes Copolymer über 80 % Propylen enthält und einen Isotaktizitäts-Index von mehr als 80 aufweist;

15 (b) von 5 bis 20 Teilen einer semi-crystallinen, im wesentlichen linearen Copolymerfraktion, die eine Kristallinität von 20 bis 60%, gemessen durch Differential-Scanning Kalorimetrie (DSC), aufweist, worin das Copolymer ausgewählt wird aus der Gruppe bestehend aus (i) Ethylen und Propylen enthaltend über 55% Ethylen, (ii) Ethylen, Propylen, und einem alpha-Olefin wie in (a) (ii) definiert, enthaltend von 1 bis 10% von dem alpha-Olefin und über 55 % von beiden Ethylen und alpha-Olefin, und (iii) Ethylen und einem alpha-Olefin, wie in (a) (ii) definiert, enthaltend über 55 % von besagtem alpha-Olefin, wobei das Copolymer bei Raum- oder Umgebungstemperatur in Xylol unlöslich ist; und

20 (c) von 40 bis 80 Teilen einer Copolymerfraktion, worin das Copolymer ausgewählt wird aus der Gruppe bestehend aus (i) Ethylen und Propylen enthaltend von 20 bis weniger als 40 % an Ethylen, (ii) Ethylen, Propylen, und einem alpha-Olefin, wie in (a) (ii) definiert, worin das alpha-Olefin in einer Menge von 1 bis 10 % zugegen ist und die Menge an vorhandenem Ethylen und alpha-Olefin von 20% bis zu weniger als 40% beträgt, und (iii) Ethylen und einem Alpha-Olefin, wie in (a) (ii) definiert, enthaltend von 20 % bis weniger als 40 % von besagtem alpha-Olefin, und gegebenenfalls mit 0,5 bis 10 % eines Diens, wobei besagte Copolymerfraktion bei Umgebungstemperatur in Xylol löslich ist und eine Grenzviscositätszahl von 1,5 bis 4,0 dl/g aufweist;

25 wobei die Gesamtmenge der Fraktionen (b) und (c), bezogen auf die gesamte Olefinpolymer-Zusammensetzung, von 50% bis 90% beträgt, und das Gewichtsverhältnis von (b)/(c) weniger als 0,4 beträgt; und

30 2) von 5 bis 40 Gew.% eines Copolymeren von Ethylen mit einem $\text{CH}_2=\text{CHR}$ alpha-Olefin, worin R ein geradkettiges oder verzweigtes C_{1-8} Alkyl bedeutet, und eine Dichte von 0,875 g/cm³ oder größer aufweist.

35

2. Das Material nach Anspruch 1, dadurch gekennzeichnet, daß (a) ein Copolymer von Propylen und Ethylen enthaltend von 90 bis 99% Propylen ist.

40 3. Das Material nach Anspruch 1, dadurch gekennzeichnet, daß (c) ein Copolymer von Propylen und Ethylen enthaltend von 20 bis 38 % Ethylen ist.

45 4. Das Material nach Anspruch 1, dadurch gekennzeichnet, daß Komponente (2) ein Ethylen-Copolymer enthaltend ein 1-Octen Comonomer ist.

50 5. Das Material nach Anspruch 4, dadurch gekennzeichnet, daß besagtes Ethylen-Copolymer eine Dichte von 0,890 bis 0,927 g/cm³ aufweist.

55 6. Das Material nach Anspruch 4, dadurch gekennzeichnet, daß besagtes Ethylen-Copolymer ein lineares Polyethylen niedriger Dichte ist.

7. Das Material nach Anspruch 1, dadurch gekennzeichnet, daß Komponente 1) in einer Menge von 90 bis 75 % anwesend ist und Komponente 2) in einer Menge von 10 bis 25 % anwesend ist.

8. Das Material nach Ansprüchen 2 oder 3, dadurch gekennzeichnet, daß der gesamte Anteil des copolymerisierten Ethylens von 15 Gew.% bis 35 Gew.% beträgt.

9. Ein Gegenstand in Form eines Films oder einer Folie enthaltend einen Basisfilm oder -folie aus einem kristallinen

Homopolymer von einem C₂-10 alpha-Olefin Monomer oder einem Copolymer ausgewählt aus der Gruppe bestehend aus (i) Propylen und mit Ethylen, (ii) Propylen mit Ethylen und einem C₄-10 alpha-Olefin Monomeren, und (iii) Propylen mit einem C₄-10 alpha-Olefin Monomeren, mit der Maßgabe, daß, falls das Comonomere Ethylen ist, die maximale polymerisierte Ethylenmenge 10 % ist, falls das Comonomere ein C₄-10 Olefin ist, die maximale davon polymerisierte Menge 20 % beträgt, und daß, falls sowohl Ethylen als auch ein C₄-10 Olefin eingesetzt werden, die maximale polymerisierte Menge 30 % beträgt; und aufgebracht auf zumindest eine Oberfläche des Basisfilms oder der -folie eine Schicht des Materials gemäß Anspruch 1.

5 10. Ein Gegenstand in Form eines Films oder einer Folie enthaltend einen Basisfilm oder -folie aus einem metallischen Substrat und darauf aufgebracht auf zumindest eine Oberfläche davon eine Schicht des Materials gemäß Anspruch 1 oder 7.

10 15. Ein Gegenstand in Form eines Nonwoven enthaltend aufgebracht auf zumindest eine Oberfläche davon eine Schicht des Materials gemäß Anspruch 1 oder 7.

Revendications

20 1. Matière en film ou en feuille comprenant un mélange constitué par 1), à concurrence de 95 à 60% en poids, une composition polymère oléfinique hétérophasique qui comprend

25 (a) de 10 à 50 parties d'un homopolymère de propylène dont l'indice isotactique est supérieur à 80, ou d'un copolymère choisi parmi le groupe constitué par (i) du propylène et de l'éthylène, (ii) du propylène, de l'éthylène et une alpha-oléfine de formule CH₂=CHR où R représente un groupe alkyle en C₂-C₈ à chaîne droite ou ramifiée, et (iii) du propylène et une alpha-oléfine telle que définie dans (ii), la teneur en propylène dudit copolymère étant supérieure à 80% et son indice isotactique étant supérieur à 80;

30 (b) de 5 à 20 parties d'une fraction de copolymère semi-cristallin essentiellement linéaire possédant une cristallinité de 20 à 60% mesurée par calorimétrie par analyse différentielle (DSC), le copolymère étant choisi parmi le groupe constitué par (i) de l'éthylène et du propylène, l'éthylène étant présent à concurrence de plus de 55%, (ii) de l'éthylène, du propylène et une alpha-oléfine telle que définie en (a) (ii) contenant l'alpha-oléfine à concurrence de 1 à 10% et à la fois l'éthylène et l'alpha-oléfine à concurrence de plus de 55%, et (iii) de l'éthylène et une alphaoléfine telle que définie dans (a) (ii) contenant ladite oléfine à concurrence de plus de 55%, ledit copolymère étant insoluble dans du xylène à la température ambiante; et

35 (c) de 40 à 80 parties d'une fraction de copolymère dans laquelle le copolymère est choisi parmi le groupe constitué par (i) de l'éthylène et du propylène contenant de l'éthylène à concurrence de 20% à moins de 40%, (ii) de l'éthylène, du propylène et une alpha-oléfine telle que définie dans (a) (ii), l'alpha-oléfine étant présente en une quantité de 1 à 10% et la quantité de l'éthylène et de l'alpha-oléfine présente étant de 20% à moins de 40%, et (iii) de l'éthylène et une alpha-oléfine telle que définie dans (a) (ii) contenant ladite alpha-oléfine à concurrence de 20% à moins de 40%, et le cas échéant un diène à concurrence de 0,5 à 10%, ladite fraction de copolymère étant soluble dans du xylène à la température ambiante, et possédant une viscosité intrinsèque de 1,5 à 4,0 dl/g;

40 45 le total des fractions (b) et (c), basé sur la composition totale du polymère oléfinique, étant de 50% à 90%, et le rapport pondéral (b)/(c) étant inférieur à 0,4; et

50 2) à concurrence de 5 à 40% en poids, un copolymère d'éthylène avec une alpha-oléfine de formule CH₂=CHR où R représente un groupe alkyle en C₁-C₈ à chaîne droite ou ramifiée, dont la masse volumique est égale à 0,875 g/cm³ ou plus.

55 2. Matière selon la revendication 1, dans laquelle (a) est un copolymère de propylène et d'éthylène contenant du propylène à concurrence de 90 à 99%.

3. Matière selon la revendication 1, dans laquelle (c) représente un copolymère de propylène et d'éthylène contenant de l'éthylène à concurrence de 20 à 38%.

4. Matière selon la revendication 1, dans laquelle le composant (2) est un copolymère d'éthylène contenant un comonomère de 1-octène.

5. Matière selon la revendication 4, dans laquelle ledit copolymère d'éthylène possède une masse volumique de 0,980 à 0,927 g/cm³.
6. Matière selon la revendication 4, dans laquelle ledit copolymère d'éthylène est un polyéthylène linéaire basse densité.
7. Matière selon la revendication 1, dans laquelle le composant 1) est présent en une quantité de 90 à 75% et le composant 2) est présent en une quantité de 10 à 25%.
- 10 8. Matière selon la revendication 2 ou 3, dans laquelle la teneur totale de l'éthylène copolymérisé représente de 15% à 35% en poids.
- 15 9. Article en film ou en feuille comprenant un film ou une feuille de base d'un homopolymère cristallin d'un monomère d'alpha-oléfine en C₂-C₁₀ ou d'un copolymère choisi parmi le groupe constitué par (i) du propylène avec de l'éthylène, (ii) du propylène avec de l'éthylène et un monomère d'alpha-oléfine en C₄-C₁₀ et (iii) du propylène avec un monomère d'alpha-oléfine en C₄-C₁₀, à condition que, lorsque le comonomère est l'éthylène, la teneur maximale en éthylène polymérisé représente environ 10%, lorsque le copolymère est une oléfine en C₄-C₁₀, sa teneur maximale à l'état polymérisé représente 20% et lorsqu'on utilise à la fois de l'éthylène et une oléfine en C₄-C₁₀, leur teneur maximale à l'état polymérisé représente 30%; ainsi qu'une couche de la matière selon la revendication 1 appliquée sur au moins une surface du film ou de la feuille de base.
- 20 10. Article en film ou en feuille comprenant un film ou une feuille de base d'un substrat métallique sur au moins une surface duquel est appliquée une couche de la matière selon la revendication 1 ou 7.
- 25 11. Article non tissé sur au moins une des surfaces duquel est appliquée une couche de la matière selon la revendication 1 ou 7.

30

35

40

45

50

55